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Magnetic Microcalorimeter Gamma Detectors for High-Precison Non-Destructive Analysis

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MAGNETIC MICROCALORIMETER GAMMA DETECTORS FOR HIGH-PRECISION NON-DESTRUCTIVE ANALYSIS

LL12-MagMicro-PD2Lb

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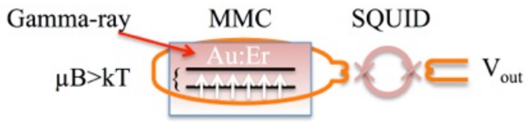
1. ABSTRACT

Cryogenic gamma (γ) detectors with operating temperatures of ~0.1 K or below offer 10× better energy resolution than conventional high-purity germanium detectors that are currently used for non-destructive analysis (NDA) of nuclear materials. This can greatly increase the accuracy of NDA, especially at low-energies where gamma rays often have similar energies and cannot be resolved by Ge detectors. We are developing cryogenic γ–detectors based on metallic magnetic calorimeters (MMCs), which have the potential of higher resolution, faster count rates and better linearity than other cryogenic detector technologies. High linearity is essential to add spectra from different pixels in detector arrays that are needed for high sensitivity. Here we discuss the fabrication of a new generation of MMC γ–detectors in FY2014, and the resulting improvements in energy resolution and linearity of the new design. As an example of the type of NDA that cryogenic detectors enable, we demonstrate the direct detection of Pu-242 emissions with our MMC γ–detectors in the presence of Pu-240, and show that a quantitative NDA analysis agrees with the mass spectrometry

2. INTRODUCTION

Metallic magnetic calorimeters (MMC) gamma detectors measure the energy of absorbed gamma rays from the resulting change in detector magnetization. Most MMC detectors currently use paramagnetic Er-dopants in a Au matrix as a sensor, a material system pioneered by Prof. Enss' group at the University of Heidelberg (Germany). A small magnetic field splits the energy levels of the spin-up and spin-down electrons due to the Zeeman effect, and at temperatures below ~100 mK the lower-energy state will be preferentially occupied. Gamma absorption will promote some of the electrons into the higher-energy state with opposite spin, and the resulting change in magnetization can be read out with a SQUID preamplifier (figure 1). This pulse signal is typically processed digitally with room temperature electronics. Its amplitude ~E/C is proportional to the energy E of the gamma ray, and its decay time $\tau_{decay} = C/G$ is set by the heat capacity C (and thus volume) of the detector and its thermal coupling G to the refrigerator.

MMCs offer three potential advantages over alternative cryogenic detector technologies. Since they do not contain any resistive elements and do not require any bias current, they have intrinsically lower noise, and can therefore offer higher energy resolution (figure 2). In addition, they are expected to offer higher linearity over a wider energy range, which is essential for adding spectra from different pixels in detector arrays. Finally, since MMCs consist of metallic Au with fewer energy traps, they are expected to decay with a single thermal decay time and thus allow the use of pulse processing algorithms that can operate under pile-up conditions and therefore at higher rates. On the other hand, MMCs are less mature than other cryogenic detector technologies, so one of the goals of this project is to determine whether MMCs can live up to their potential or whether some non-idealities limit their performance.



$$\Delta E_{FWHM} \approx 2.355\sqrt{4kT^2C \cdot 2\sqrt{\frac{\tau_{rise}}{\tau_{decay}}}}$$

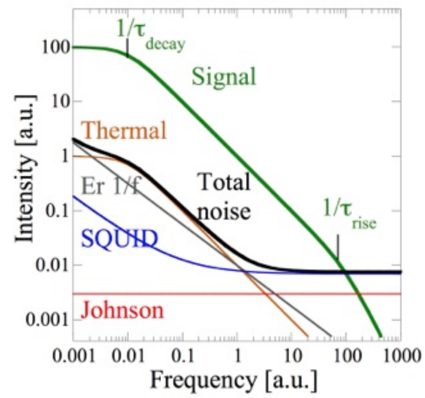


Figure 1 (left): MMC schematic. Gamma absorption promotes electrons into the higher-energy state with opposite spin, and the resulting change in magnetization is read out with a SQUID preamplifier. The equation shows the limiting energy resolution for an MMC with heat capacity C and operated at temperature T. Figure 2 (right): MMC noise characteristics. The white noise is set not by the MMC, but by the SQUID amplifier.

The goal of this project is therefore to develop ultra-high energy resolution γ-detectors based on magnetic micro-calorimeters (MMCs), and to determine whether these detectors live up to their potential experimentally. For this, we are collaborating with Prof. Christian Enss at the University of Heidelberg, the world's leading group in the field of MMC detector development. Specifically, we are adapting the MMC X-ray detector design from Heidelberg for higher-energy gamma-ray detection. Since MMCs, like other cryogenic γ-detector technologies with operating temperatures <0.1 K, are intrinsically slow and have to be small for high energy resolution, we will place an emphasis on questions that determine sensitivity and the potential for scaling to arrays. The objectives are therefore to fabricate an improved version of MMC γ-detectors and test their energy resolution, maximum count rate, readout noise, crosstalk between pixels and linearity.

3. MMC DETECTOR FABRICATION

Earlier MMC γ-detectors were fabricated by depositing the Au gamma absorber directly onto the magnetic Au:Er sensor (figure 3). Their energy resolution was limited by varying amounts of energy loss into the substrate, especially at the higher energies relevant for safeguards NDA. In FY14, we have sent our graduate student Cameron Bates to work with our collaborators at the University of Heidelberg and fabricate the next generation of MMC gamma detectors. In the new design, the absorber is supported on Au posts, so that the gamma energy is fully thermalized in the Au absorber before reaching the Au:Er sensor (Figure 4). This was expected to reduce the line broadening from partial energy loss into the Si substrate. For this, Cameron had to develop a new lithographic process that involved patterning thick AZ125nXT photoresist into a 200 µm deep mold for electroplating the Au absorber onto the posts.

As in the earlier design, the input to the SQUID preamplifier consist of two coils in a gradiometer configuration with different polarity windings, so that each sensor has two pixels that produce signals with opposite polarity. While the gradiometer design increases the electronic noise by a factor of √2, it also increases the number of pixels and makes the MMC less susceptible to pick up electromagnetic interferences. The new fabrication process was successful, and the first wafer has already improved the MMC device yield from <10% to ~50%.

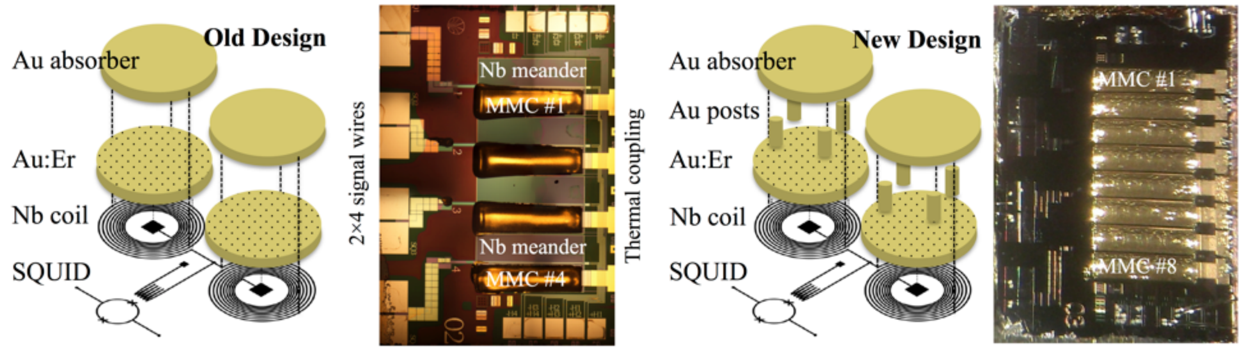


Figure 3a/3b (left): In the earlier MMC design, the gold absorber was directly deposited onto the Au:Er sensor (3a). Its size was poorly defined and it overlapped with the next pixel so that only 4 of the 8 pixels were usable (3b). Figure 4a/4b (right): In the MMC gamma detectors we built in FY14, the absorber is supported by Au posts, so that the gamma energy can thermalize completely in the absorber before heating the Au:Er sensor (4a). In addition, we improved the photolithographic definition of the absorber so that all 8 pixels of the arrays can now be used (4b).

4. REFRIGERATION

We have optimized our liquid-cryogen-free adiabatic demagnetization refrigerator (ADR) so that the MMCs can now be cooled to a base temperature <35 mK. This required better thermal shielding around the MMC detector, and some aluminum tape to close light leaks in the higher-temperature shields that had increased the heat load into the detector stage. For lowest temperature operation and longest hold time, we also add more helium gas to the compressor of the pulse-tube cryo-cooler, so that its precool temperature is reduced from 3.2 to 2.7 K, which subsequently lowers the ADR base temperature. We found that we can fill the compressor to its maximum design pressure of He only after the pulse tube has been initially cooled below 5 K, otherwise the power consumption during cool-down trips the compressor's circuit breakers. Our ADR does not require any cryogenic liquids, and its cool-down is fully automated.

While a base temperature of <35 mK should be sufficient for the goals of this proposal, namely to attain an energy resolution <100 eV, our ADR requires periodic cycling to 4 K every ~12 hours, which makes long data acquisition from weak radioactive source cumbersome. We have therefore started a collaboration with Peter Shirron at NASA Goddard to build a continuous adiabatic demagnetization refrigerator (CADR) that can keep MMC gamma detectors at a temperature of ~35 mK indefinitely. The CADR contains four independent magnets with four separate paramagnets that go through the demagnetization cycle periodically in such a fashion that the temperature of the lowest stage can be kept cold continuously. To precool the CADR from room temperature to ~3 K, we have chosen a comparably low-power Gifford-McMahon (GM) cryo-cooler. While GM coolers tend to be noisier than pulse tube refrigerators, they are also more efficient, and consequently can be run on regular 110 V line power and do not require cooling water. The CADR can therefore be made comparably compact and integrated into an instrument that can be put on wheels to make it transportable. If the increased noise of the GM cooler turns out to preclude high-resolution MMC operation, the instrument is designed so that the GM cooler can be replaced with a lower-noise pulse tube refrigerator (with the attendant need for cooling water and 280 V, and loss in transportability).

The continuous cooling power of the CADR will enable long-term automated data acquisition and NDA from weak sources, and the transportable design will allow moving the MMC to nuclear sources that cannot be transported or into laboratories of collaborators.

5. GAMMA SPECTROSCOPY RESULTS, DISCUSSION AND CONCLUSIONS

5.1 SPECTRA FROM HEIDELBERG

The improved detector fabrication process has directly led to a significant improvement in energy resolution. We have tested the new MMCs with our collaborators at the University of Heidelberg in their dilution refrigerator with a base temperature of 14 mK. The gamma rays from the Am-241 source were collimated onto the MMC detector with a Pb pinhole, the full signal waveforms were captured and processed with an optimal filter. The measured energy resolution of 46 eV FWHM is the best resolution ever obtained with an MMC gamma detector (figure 5). As expected, the line broadening at higher energies is greatly reduced due to full thermalization of the gamma energy in the Au absorber. In addition, the thermalization improved the linearity of the detector response, which we had to study in more detail at LLNL. The question then became which energy resolution these detectors would be able to achieve at LLNL, where we currently do not have a dilution refrigerator and are therefore limited to our ADR base temperature of ~31 mK.

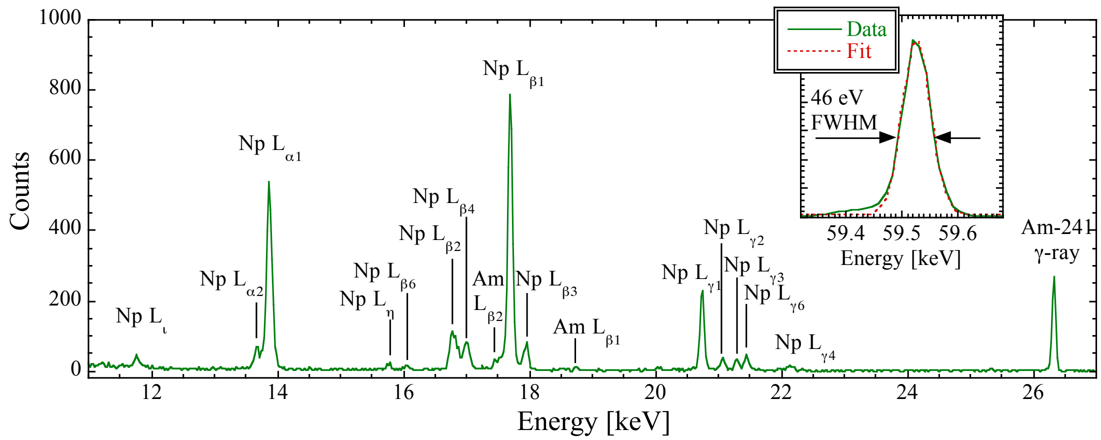


Figure 5: High-resolution Am-241 spectrum taken with one of our new MMC gamma detectors in a dilution refrigerator at a temperature of 14 mK at the University of Heidelberg. Note the large numbers of minor X-ray transitions that could never be resolved with a high-purity Ge detector.

5.2 SPECTRA FROM LLNL

We have repeated the measurements at LLNL in our ADR cryostat with a base temperature of only ~31 to ~35 mK, which significantly decreases the signal height and therefore the energy resolution. Figure 6 shows the detector response to a Pu-242 source. We have selected this source for the initial demonstration experiment, because the direct detection of Pu-242 in mixed-isotope Pu samples, which it cannot be done with Ge detectors, would be of great interest for the safeguards community. As in the Heidelberg setup, the full MMC signal waveforms were captured and filtered digitally. During the detector testing, we have taken 32 spectra from the two pixels of our MMC gamma detector, calibrated them linearly and added them with 20 eV bins. The energy resolution of this combined spectrum is 150 eV FWHM, although individual spectra have a resolution as good as 130 eV FWHM at 60 keV. We will continue to work on understanding the sources of noise and the variations in energy resolution in FY15.

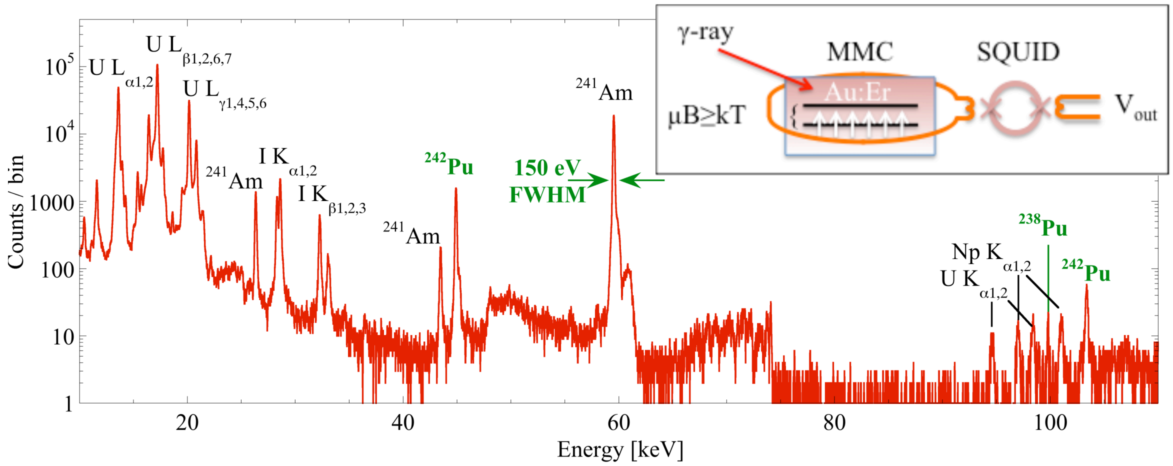
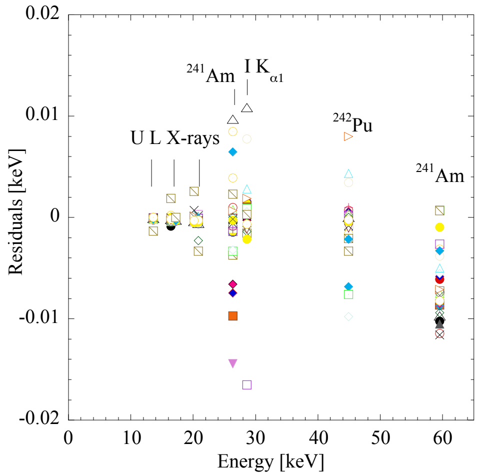


Figure 6: Sum of 32 spectra from a Pu-242 source, taken with the same MMC gamma detector as above in our ADR cryostat at LLNL at a higher temperature of ~35 mK.

More importantly, the response of the MMC gamma detector is very linear. This is crucial because adding the response from different pixels is essential for cryogenic detectors, because the individual pixels have to be very small and their sensitivity can therefore only be increased by fabricating detector arrays. For example, the 32 individual spectra that contributed to figure 6 were calibrated using only the strong lines below 60 keV. Still, after summing the spectra, the weaker lines in the 100 keV region emerge with the _same_ high energy resolution as the low-energy lines, indicating a high level on linearity (figure 6). This is also demonstrated in the plot of the residuals to the linear fit, which are below 10 eV (Figure 7). In addition, the individual spectra were acquired during separate and independent cool-downs. While the operating conditions of the MMC during these runs were nominally identical, it is very reassuring that the detector response is extremely reproducible between different runs. This result is not as trivial as it may sound initially. In the past, we have never been able fabricate transition edge sensor (TES) microcalorimeter gamma detectors with the same level of linearity and reproducibility. As a consequence, the TES energy resolution always decreased when adding spectra from different pixels and different cool-downs. The new result suggests that scaling MMCs to arrays for higher sensitivity and thus faster and higher-accuracy NDA should be very much doable, and probably more easily than with TES gamma detectors. These initial results provide experimental evidence for one of the anticipated advantages of MMC gamma-detectors, namely their increased linearity and reproducibility.

One limitation of the current MMC design is its susceptibility to develop shorts between the Au cooling pad of the MMC detector and the superconducting Nb coil that is used to apply a magnetic field for the Zeeman splitting of the energy levels in Er. While we handled the MMC detectors extremely carefully to avoid electrostatic discharges, we have not been able to avoid these shorts and the associated loss of the detector. Neither have our collaborators in Heidelberg. In particular, we have learned (through painful experience) that MMCs that had initially seemed functional tend to exhibit shorts after wire bonding. This suggest that these shorts occur either because of an electrostatic discharge during bonding (Heidelberg's hypothesis), or because of stress in the insulator between the Au wire to the cooling pad and the underlying Nb coil, which causes the insulator to crack, either immediately or during cool-down (LLNL hypothesis). This problem will be addressed in the next MMC design by re-routing the Nb coil wires.



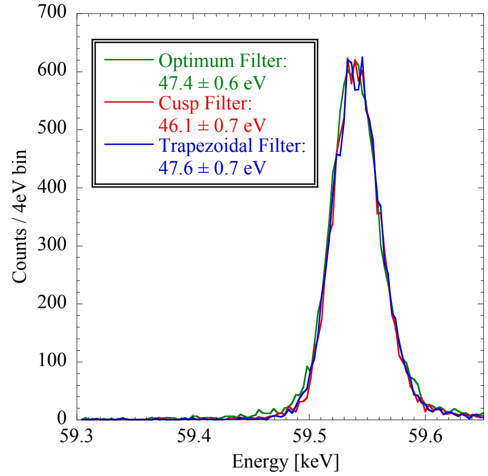


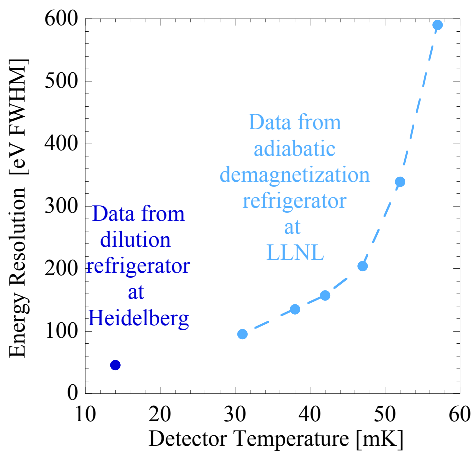
Figure 7 (left): The residuals to a linear fit of the 32 spectra that went into figure 2 are extremely small. Figure 8 (right): Our trapezoidal filter routine produces the same high energy resolution with the Heidelberg data from figure 5 as the optimum filter (which is only "optimum" for identical signal shapes and stationary noise).

One consequence of the shorts in the MMCs is that we have not yet been able to add wires from the Au thermalization pads to the cryostat cold plate in order to improve the thermal coupling of the MMCs and speed up their response. (These pads are on the right hand side of the pictures in figure 3b and 4b.) This approach has been used successfully for the MMC X-ray detectors at Heidelberg, and is responsible for their single MMC decay constant of ~1 ms. In contrast, our MMCs currently have decay constants >10 ms, because the MMCs have to cool through the Si substrate and the varnish that glues the chip to the cryostat cold plate. In addition, the signals exhibit a small secondary decay time that is even longer, probably due to a small amount of stored energy in some dielectric part of the chip, and similar to what is affecting the response of TES gamma detectors. This has kept us so far from operating the MMCs above rates of a few counts per second. We do believe that this problem can be fixed in the next generation of MMCs when we should be able to use Au bond wires to improve MMC cooling.

As part of our data acquisition development during FY15, we have also developed several digital filtering routines to determine which algorithm produces the highest energy resolution with MMC signals. (MMC require custom pulse processing routines because a single SQUID outputs signals with both polarities from the two gradiometer coils.) We have tested these digital filters with the high resolution data we took in Heidelberg (see Figure 5), and compared them to the "optimum filter" results of our Heidelberg collaborators. An optimum (Wiener) filter constructs the filter function based on the measured spectral characteristics of the signal and the baseline noise, by minimizing the chi-squared between each waveform and a noise-free waveform template. However, such an optimum filter assumes that the normalized signal shape and the underlying noise are constant throughout a run, which may not be true to the degree needed, in which case the energy resolution can be reduced. Under these circumstances, cusp filters and trapezoidal filters may produce equally high or even higher energy resolution. The results in Figure 8 suggest that our cusp and trapezoidal filters work well, and sometimes even better than the optimum filter. We are therefore in a good position to test the faster response of the next generation MMCs in FY15.

5.3 FUTURE WORK

For safeguards and other NDA applications, the primary interest in MMC is due to their exquisitely high energy resolution, since it enables NDA on certain isotopes that are not measureable with Ge detectors. The second goal is then to perform NDA as quickly as possible without loss in energy resolution. Finally, a practical instrument must be accessible to non-expert users, and its operation should therefore be automated if at all possible to reduce cost.



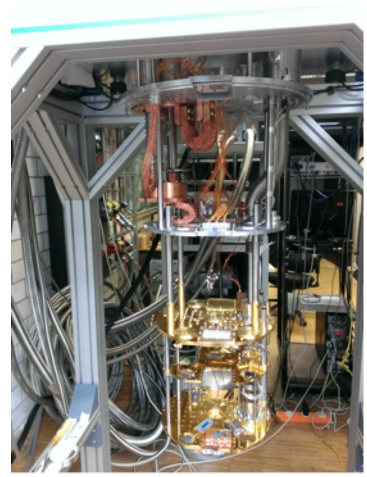


Figure 9 (left): The energy resolution of our MMC improves as the temperature is lowered as expected, primarily because the signal increases roughly proportional to 1/T (Curie law). Figure 10 (center): The BlueFors dilution refrigerator at Heidelberg has a base temperature of <15 mK, and its operation is liquid-cryogen-free and fully automated. Figure 11 (right): The BlueFors refrigerator is designed such that it can be disassembled to install new detectors without lowering its outer shell into a hole in the ground, a significant improvement over older designs.

The path forward should therefore focus on improving the performance of single pixel MMC gamma detectors, and on developing arrays of these MMCs to improve sensitivity. For improved energy resolution, a dilution refrigerator with a base temperature <15 mK would be very desirable, as shown by comparing our 14 mK results from Heidelberg with our >30 mK data from LLNL (Figures 9). Although we knew that a dilution refrigerator would further improve the energy resolution of MMCs, we had not requested funds for this comparably expensive instrument in this proposal because it was not clear whether we could build MMC gamma detectors with the necessary energy resolution, linearity and reproducibility. Our results from FY14 show that this is the case, making the purchase of a dilution refrigerator very desirable. If a dilution refrigerator with pulse-tube pre-cooling is used, like the BlueFors refrigerator in Heidelberg, it would provide the additional benefit that the operation would be fully automated, and that the MMC detectors could be held at their 15 mK operating temperature indefinitely.

For improved detector speed and thus faster NDA analysis, a better coupling of the MMC detector to the cryostat base temperature is essential. This will be accomplished with a re-design of the thermalization path to increase the thermal coupling between the MMC and the cryostat cold stage using Au wire bonds. While the metallic thermal path should make this possible, it remains to be confirmed experimentally that such a design would in fact speed up the MMC gamma detector response to ~1 ms, as it does for the MMC X-ray detectors. If MMCs decay with a single decay time, count rates of ~100 counts/s per pixel should be possible.

For improved sensitivity, detector arrays must be developed. Initially, these arrays should be of moderate size arrays to test and optimize thermal coupling, cross-talk and readout. But eventually, it would be very desirable to develop large arrays with several thousands of MMCs to reduce the time for NDA with MMC. This will require some form of multiplexing technology to read out many pixels with few wires, because one cannot bring thousands of wires to the cold stage of a ~15 mK refrigerator without heating it up. Since the multiplexing technology for MMC detectors is not very mature (yet), the arrays readout should initially be pursued by parallel readout of individual MMCs.

While DOE may not want to invest the millions of dollars required for developing a reliable multiplexed readout, this is of great importance for NASA and other space agencies that want to image extended astronomical X-ray sources. For this reason, there are already efforts at Heidelberg / Magnicon and at NASA Goddard / NIST to develop microwave multiplexers for MMC readout. The basic concepts have been demonstrated, and the detailed engineering and optimization efforts are continuing. We consider it desirable for DOE to stay one step behind the astronomer's efforts, and collaborate with them on the adaptation of X-ray multiplexers for MMC gamma detectors, because the potential for fully automated NDA of nuclear materials with large arrays of high-resolution MMCs is very high.

6. PUBLICATIONS

"Development of MMC Gamma Detectors for Nuclear Analysis", C. R. Bates, C. Pies, S. Kempf, L. Gastaldo, A. Fleischmann, C. Enss, S. Friedrich, *J. Low Temp. Phys*. **176** (2014)